

# Tunnel Spin Polarization Versus Energy for Clean and Doped $\text{Al}_2\text{O}_3$ Barriers

B. G. Park, T. Banerjee, J. C. Lodder, and R. Jansen

*MESA<sup>+</sup> Institute for Nanotechnology, University of Twente, 7500 AE Enschede, The Netherlands*

(Received 20 January 2007; published 20 November 2007)

The variation of the tunnel spin-polarization (TSP) with energy is determined using a magnetic tunnel transistor, allowing quantification of the energy dependent TSP separately for both ferromagnet/insulator interfaces and direct correlation with the tunnel magnetoresistance (TMR) measured in the same device. The intrinsic TSP is reduced below the Fermi level, and more strongly so for tunneling into empty states above the Fermi level. For artificially doped barriers, the low bias TMR decreases due to defect-assisted tunneling. Yet, this mechanism becomes ineffective at large bias, where instead inelastic spin scattering causes a strong TMR decay.

DOI: [10.1103/PhysRevLett.99.217206](https://doi.org/10.1103/PhysRevLett.99.217206)

PACS numbers: 85.75.-d, 73.40.Gk, 75.70.-i

A magnetic tunnel junction (MTJ) is of great interest owing to its large tunnel magnetoresistance (TMR) [1–3], which is applied in spin-based electronic devices such as magnetic random access memories, magnetic recording read heads, and spin torque devices [4–6]. In a MTJ consisting of two ferromagnetic (FM) layers separated by a thin insulator, the tunneling resistance depends on the relative orientation of the magnetization of the FM electrodes due to the tunnel spin polarization (TSP) associated with the FM/insulator interfaces [7]. A characteristic is that the TMR is significantly reduced at larger bias voltage [8]. Proposed explanations include magnon excitation at the FM/insulator interface [9,10], defect mediated tunneling [11,12], and intrinsic electronic structure effects [13,14]. At small bias where the largest TMR is obtained, only states near the Fermi level ( $E_F$ ) are involved in the tunneling process. At a large bias, there is a substantial contribution of electrons which tunnel from the occupied states below  $E_F$  of one electrode and/or tunnel into the empty states above  $E_F$  of the other electrode, thereby altering the TMR. Also, when a large bias (current) is applied to a MTJ, excitation or even switching of the magnetization may occur due to a spin-transfer torque [15].

To understand the bias reduction of the TMR as well as the spin-transfer torque of a MTJ, it is crucial to determine the TSP versus energy, and separately for the filled states of one electrode and the empty states of the other electrode. Moreover, it is of equal importance to examine the role of barrier defects. Studies of the TSP of different FM/insulator combinations have mostly been done using spin-polarized tunneling into a superconducting counter electrode [7], which probes the TSP of the states near  $E_F$ . Recently, the first direct measurements showing that the TSP decreases with increasing bias were reported [16], using a lateral mesoscopic spin-valve structure of FM/ $\text{Al}_2\text{O}_3$ /Al strip/ $\text{Al}_2\text{O}_3$ /FM. In this approach, there is no FM/ $\text{Al}_2\text{O}_3$ /FM junction to determine the TMR, and thus no direct correlation between TSP and the voltage dependence of the TMR can be made. Also, the barrier quality cannot be judged from the TMR and its decay with

bias, while quantification of the TSP requires the values of the spin diffusion parameters of the Al strip to be measured [17].

In this Letter, we report on the determination of the energy dependence of the TSP using a magnetic tunnel transistor (MTT), which combines a MTJ with an  $n$ -type Schottky diode [18] (see inset of Fig. 1). The magnetic response of the MTT is shown to allow quantitative probing of the TSP versus energy of the emitter FM/insulator interface. The TSP of the other (base) interface is obtained independently from the correlation with the TMR measured in the same device. The TSP is found to be largest at  $E_F$  and most strongly reduced above  $E_F$ . In addition, using artificially doped tunnel barriers, we demonstrate that barrier defects affect the TSP at low and high bias via different mechanisms.

MTT's of  $(\text{Ni}_{80}\text{Fe}_{20} \text{ or } \text{Co})/\text{Al}_2\text{O}_3/\text{Co}/\text{Au}/n\text{-type Si}$  were fabricated as previously described [18]. A Schottky barrier of  $\sim 0.8$  eV was formed at the Au/ $n$ -type Si interface. Transport measurements were conducted at 100 K using a four-point geometry for the emitter to base tunnel junction and a separate ohmic contact to the Si collector. In a MTT with a FM emitter, spin-polarized hot electrons are injected into the FM (Co) base by tunneling. After spin-dependent transmission through the FM base, electrons are collected in the conduction band of the  $n$ -type Si if they have retained proper energy and momentum to overcome the Au/Si Schottky barrier. The magnetic response of the MTT, the so called magnetocurrent (MC), is determined by the  $\text{TSP}_E$  of the emitter interface and the spin-asymmetry of the base transmission ( $T_B$ ), and expressed [19] as  $\text{MC} = 2 \times \text{TSP}_E \times T_B / (1 - \text{TSP}_E \times T_B)$ . Throughout the manuscript, we use the term TSP to denote an effective tunnel spin-polarization, which depends on the electronic structure of the FM/insulator combination, the tunneling matrix elements, energy, as well as temperature, and thus not just on the density of states of the FM. The TMR can then be factorized in terms of the  $\text{TSP}_E$  and  $\text{TSP}_B$  of the emitter and base interfaces, respectively, using  $\text{TMR} = 2 \times \text{TSP}_E \times \text{TSP}_B / (1 - \text{TSP}_E \times \text{TSP}_B)$ . This has been justified

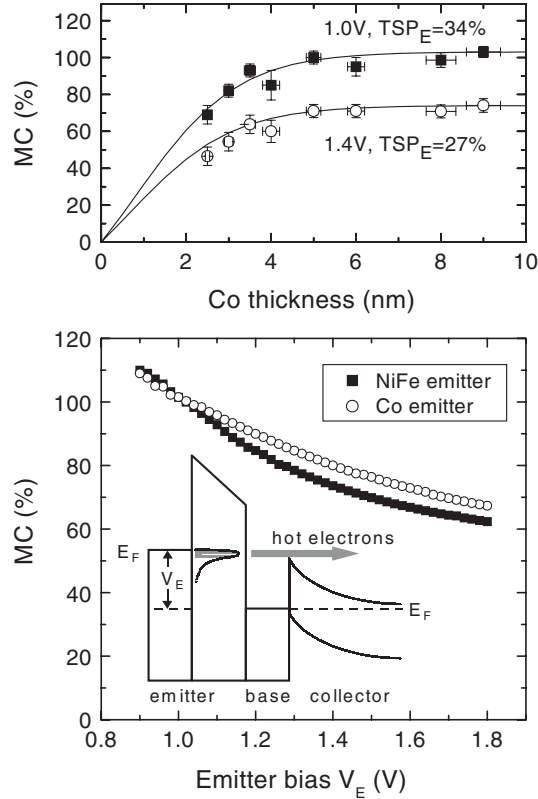


FIG. 1. Top panel: magnetocurrent (MC) as a function of the Co base thickness at an emitter bias of 1.0 and 1.4 V for an MTT of Ni<sub>80</sub>Fe<sub>20</sub>/Al<sub>2</sub>O<sub>3</sub>/Co/Au/*n*-type Si. The solid lines are fits for determination of the spin-asymmetry in the base attenuation lengths ( $\lambda_M/\lambda_m$ ). Bottom panel: MC as a function of emitter bias for an MTT with Co or Ni<sub>80</sub>Fe<sub>20</sub> emitter.  $T = 100$  K. Inset is the schematic energy diagram of the MTT, with the gray area indicating the energy states contributing to the MC near the collection threshold.

by theory [20] as well as experiments; for example, the TMR of junctions with amorphous Al<sub>2</sub>O<sub>3</sub> was shown to be consistent with the TSP of each electrode determined separately [10]. Below we show that this also applies to the MTJ's used here.

The top panel of Fig. 1 shows the MC versus Co base thickness ( $t_{Co}$ ) at an emitter bias of 1.0 and 1.4 V, where the emitter FM is Ni<sub>80</sub>Fe<sub>20</sub>. Here, the MC is measured as  $(I_C^P - I_C^{AP})/I_C^{AP}$ , where  $I_C^{P(AP)}$  is the collector current for parallel (antiparallel) alignment of emitter and base FM. As  $t_{Co}$  is increased, the MC initially increases, but saturation occurs at  $t_{Co} \sim 5$  nm. Since the TSP of the emitter FM is not varied with  $t_{Co}$ , the initial increase is due to the enhancement of the spin-filtering effect of the Co base as determined by the spin-dependent hot-electron attenuation lengths  $\lambda_M$  and  $\lambda_m$  for majority and minority spins, respectively [19]. The  $\lambda_M$  value extracted from the exponential decay of  $I_C^P$  versus  $t_{Co}$  is  $\sim 7$  nm (not shown). Using this value in a fit (solid lines) of the MC versus  $t_{Co}$ , we obtain  $\lambda_m \sim 1$  nm. The saturation MC of a MTT with a 9 nm thick Co base is 103% for a bias voltage of 1.0 V,

while the MC is only 73% when the bias voltage is raised to 1.4 V. The saturation of the MC for thick base occurs because only majority spin electrons are transmitted by the Co, resulting in a spin-asymmetry of the base transmission of unity (for  $t_{Co} = 9$  nm and the attenuation lengths given above,  $T_B$  is  $+0.999$  and positive since  $\lambda_M > \lambda_m$ ). Consequently, for a thick Co base, the MC is only dependent on the TSP of the emitter interface  $[MC = 2 \times TSP_E/(1 - TSP_E)]$ . Hence, the lower MC for larger voltage must be due to the dependence of the TSP on the bias voltage. The resultant TSP<sub>E</sub> at an emitter bias of 1.0 V (1.4 V) is 34% (27%) and positive, as expected [7,10]. The bottom panel of Fig. 1 shows the MC for MTT's with a Co and Ni<sub>80</sub>Fe<sub>20</sub> emitter as a function of the emitter voltage, with an 8 nm thick Co base. For an emitter bias of 0.9 V, which is just above the Schottky barrier height (0.8 eV) of the Au/Si collector diode (see inset), the MC is 110%. The MC decreases monotonously with increasing emitter voltage, and slightly weaker so for the MTT with a Co emitter.

The TSP<sub>E</sub>(V) values extracted from the MC at each bias are shown in the top panel of Fig. 2 for a MTT with a Co emitter, where the horizontal scale is the voltage with respect to the threshold ( $V_E - 0.8$  V). The TSP<sub>E</sub> is 35%

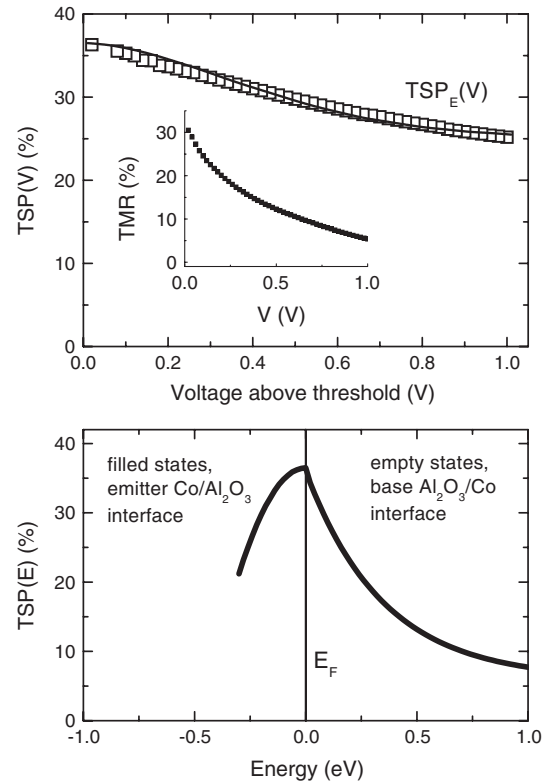


FIG. 2. Top panel: effective TSP<sub>E</sub>(V) versus bias voltage for the emitter Co/Al<sub>2</sub>O<sub>3</sub> interface, extracted from the MC. Inset is the TMR versus bias voltage of the emitter to base Co/Al<sub>2</sub>O<sub>3</sub>/Co tunnel junction. Bottom panel: extracted TSP versus energy with respect to  $E_F$  at 100 K. Convolution with the energy distribution of tunnel electrons gives the solid line in the top panel.

at 0.1 V above the threshold and decreases to 25% at 1.0 V. For a bias near the 0.8 eV collection threshold, only electrons tunneling from the states near  $E_F$  of the emitter can overcome the Schottky barrier (see inset of Fig. 1). Therefore, the TSP at the threshold bias is associated with the filled states near  $E_F$  of the Co/Al<sub>2</sub>O<sub>3</sub> emitter interface. For larger bias, electrons tunneling from states below  $E_F$  of the Co/Al<sub>2</sub>O<sub>3</sub> emitter can also contribute to the collector current. Thus, the reduced TSP for larger bias above the threshold implies that the TSP is reduced below  $E_F$  of the Co/Al<sub>2</sub>O<sub>3</sub> emitter. From the TSP<sub>E</sub>(V) data, we extract the TSP versus energy below  $E_F$ . Assuming there is no spin-flip scattering during tunneling, we have TSP<sub>E</sub>(V)  $\propto$   $\int_0^V$  TSP<sub>E</sub>(E)  $I_T$ (E) dE, where TSP(E) is for states with energy E with respect to  $E_F$ , and  $I_T$ (E) is the normalized energy distribution of the tunnel current, taken to be the same for both spins. This implies that any spin dependence (for example, due to a spin-dependent wave vector) is incorporated into TSP(E). In a free-electron model,  $I_T$ (E) can easily be calculated [21], using experimental parameters for the tunnel barrier width ( $1.8 \pm 0.2$  nm) and height ( $2.5 \pm 0.3$  eV, determined from a fit [22] of the  $dI/dV$  vs V curve). The bottom panel of Fig. 2 shows the extracted TSP(E) of the emitter Co/Al<sub>2</sub>O<sub>3</sub> interface versus energy (data for  $E < E_F$ ). The TSP at  $E_F$  is largest (36.6%) and is reduced below  $E_F$ . The TSP at energy more than 0.3 eV below  $E_F$  cannot be determined accurately because of the negligible contribution to the tunneling, since the energy distribution of the tunneling current is peaked near  $E_F$  of the emitter [21].

The same MTT can be used to obtain the TMR versus bias voltage of the emitter to base Co/Al<sub>2</sub>O<sub>3</sub>/Co tunnel junction, as shown in the inset of Fig. 2. The TMR is 31% at a bias voltage of 20 mV. Using  $\text{TMR} = 2 \times \text{TSP}_E \times \text{TSP}_B / (1 - \text{TSP}_E \times \text{TSP}_B)$ , this corresponds to a TSP of 36.4% at  $E_F$  for both Co/Al<sub>2</sub>O<sub>3</sub> interfaces. This is consistent with the TSP value for the emitter interface extracted independently from the MC, proving that factorization is justified for our junctions. This was also found in previous experiments [10] and expected from theory [20], in which this was shown to be justified, amongst others, when the tunnel barrier and/or interfaces are sufficiently disordered, which applies to our junctions with amorphous Al<sub>2</sub>O<sub>3</sub> barriers and polycrystalline, nonepitaxial electrodes. The TSP extracted from the low bias TMR (where receiving states are near  $E_F$  of the base) and the TSP extracted from the MC (where receiving states are at least 0.8 eV above  $E_F$  of the base) are in agreement, i.e., the TSP at  $E_F$  of the emitter Co/Al<sub>2</sub>O<sub>3</sub> interface is independent of the final states of the base counter electrode, supporting the validity of the factorization over a wide bias range.

Next, we extract TSP<sub>B</sub>(E) of the empty states above  $E_F$ , recognizing that the TMR at bias V is determined by TSP<sub>E</sub>(E) below  $E_F$  of the emitter, the TSP<sub>B</sub>(E) up to eV above  $E_F$  of the base, and an integration over energy using the energy distribution  $I_T$ (E) of tunnel electrons:

$\text{TMR}(V) = \int_0^V [2\text{TSP}_E(E - V)\text{TSP}_B(E)] I_T(E) dE / \int_0^V [1 - \text{TSP}_E(E - V)\text{TSP}_B(E)] I_T(E) dE$ . Using the measured TMR(V), the TSP<sub>E</sub> values for states below  $E_F$  already extracted from the MC, and the same  $I_T$ (E) as used before, the TSP<sub>B</sub>(E) is then obtained. The result is shown in the bottom panel of Fig. 2 for  $E > E_F$ . Compared to energies below  $E_F$ , the TSP above  $E_F$  is reduced more strongly. This was also found in Ref. [16], but we do not observe the sign reversal of the TSP at large bias. It is difficult to understand the origin of this difference as no data on the TMR and its decay with V for the junctions used in Ref. [16] is available for comparison.

The decay of the TSP for states away from  $E_F$  can be related to the energy dependence of the electronic structure of the Co/Al<sub>2</sub>O<sub>3</sub> interface [14], and/or spin-wave excitations by tunneling electrons at the electrode/barrier interfaces [9,10], where the latter have a typical energy below about 150 meV. However, if the tunnel barrier contains defects, resonant tunneling or spin flip scattering due to inelastic scattering via defects can occur [11,12,23,24], degrading the TMR. Hence, we examine the effect of defects in the tunnel barrier on the energy variation of the TSP using defects artificially introduced in the Al<sub>2</sub>O<sub>3</sub> tunnel barrier of an MTT by means of a small amount of Cu-ion doping. The Cu-doped Al<sub>2</sub>O<sub>3</sub> tunnel barrier was formed by a double step process [11] in order to avoid any possible relocation of the barrier materials during the oxidation. First, an Al<sub>2</sub>O<sub>3</sub> barrier was formed by plasma oxidation of an Al layer of 0.9 nm. Then, Cu of less than a monolayer (0.4 to 1.2 Å) followed by a second Al layer of 0.8 nm was deposited and oxidized.

The TMR at low bias (20 mV) as a function of Cu content is shown in the top panel of Fig. 3. The TMR is 32% for a clean Al<sub>2</sub>O<sub>3</sub> barrier. As the amount of Cu doping increases, the TMR is gradually reduced and becomes  $\sim 16\%$  for a barrier doped with 1.2 Å of Cu. Figure 3, bottom panel, shows the MC as a function of the emitter bias for MTT's with Cu-doped Al<sub>2</sub>O<sub>3</sub> barriers. Independent of the amount of Cu doping, the MTT's show a MC approaching 110% at the threshold voltage of 0.8 V. The MC near the threshold is thus insensitive to the defect concentration in the tunnel barrier. This is surprising given that the TMR at low bias (top panel) is significantly reduced. A second observation is that the MC for MTT's with Cu-doped barriers decays much more strongly with increasing bias voltage as compared to the MTT with a clean Al<sub>2</sub>O<sub>3</sub> tunnel barrier. The larger the Cu content, the stronger the bias voltage dependence.

The MTT data allows us to separate elastic from inelastic scattering of the tunnel electrons by the Cu dopants. At the threshold, only electrons tunneling from the states at  $E_F$  of the emitter can contribute to the collector current, and only if there is no energy loss in the tunneling process. The MC value at the collection threshold of the MTT's is hardly affected by the Cu, even though (quasi-)elastic scattering processes must be present as evidenced by the

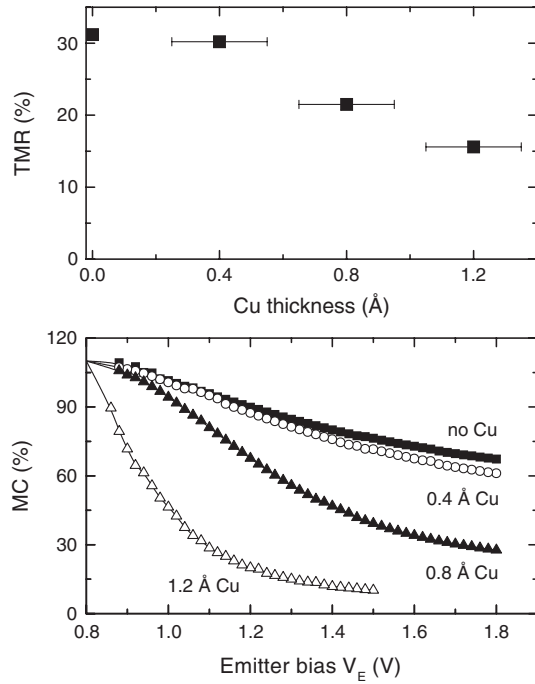


FIG. 3. Top panel: TMR (at 20 mV) versus Cu content measured in MTT devices with Cu-doped tunnel barriers. Bottom panel: MC as a function of emitter bias for the same MTT devices, with Cu content as indicated. The lines are a guide to the eyes.  $T = 100$  K.

reduction of the TMR at low bias (Fig. 3, top panel), which implies that the effective TSP at  $E_F$  is reduced. Therefore, a lower MC at the collection threshold of Cu-doped MTT's was expected, but not observed. The apparently contradictory results can be understood by tunneling via dopant-induced localized states in the barrier, if the different energy (and thus spin-polarization) of the receiving states of the Co base is taken into account. At the low bias of the TMR measurement, these localized states couple with the highly-polarized states near  $E_F$  of both the FM electrodes, producing a defect-assisted tunnel current with reduced spin-polarization due to the admixture of the unpolarized defect states [23]. This explains the reduced TMR. For the MTT at the threshold, the same states near  $E_F$  of the emitter now couple to empty states of the receiving Co base electrode that are far ( $\sim 0.8$  V) above  $E_F$ . Since these states already have a very weak tunnel spin-polarization (see Fig. 2, bottom panel), the admixture of the unpolarized defect states has little further effect. This explains why the MC of the MTT at the threshold voltage is rather insensitive to the defect concentration in the barrier. Thus, we find that (quasi-)elastic defect-assisted tunneling affects the TSP mainly at low bias.

Since barrier defects are known [11,12] to enhance the bias dependence of the TMR, the responsible processes must be inelastic in nature. This picture is indeed confirmed by the data for Cu-doped MTT's at larger emitter

voltages. In this case, electrons originating from emitter states far above the threshold can experience inelastic spin-exchange scattering processes in the tunnel barrier and still retain sufficient energy to be collected. The spin scattering reduces the spin polarization of the tunnel current and thereby the MC. Thus, the MC well above the threshold reflects the intrinsic TSP of the emitter interface reduced by spin scattering in the barrier. The strong effect of Cu-doping in this situation (see Fig. 3) shows that the TSP at large bias is very sensitive to inelastic spin scattering by defects in the tunnel barrier. Given this, it would be of interest to investigate how such defects affect the spin-transfer torque in MTJ's [15].

We acknowledge financial support from the NWO VIDI program and NanoNed, a national nanotechnology program coordinated by the Dutch Ministry of Economic Affairs.

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